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QUASI INSTANTANEOUS REACTION IN NONHOMOGENEOUS TURBULENT  
FIELD. INFLUENCE OF THE DISTRIBUTION OF TURBULENT ENERGY  
DISSIPATION ON CONVERSION RATE

M. Fenoglio, M. Pipino, S. Valerio and A. A. Barresi

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# ABSTRACTS

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# QUASI INSTANTANEOUS REACTION IN NON HOMOGENEOUS TURBULENT FIELD INFLUENCE OF THE DISTRIBUTION OF TURBULENT ENERGY DISSIPATION ON CONVERSION RATE

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## Introduction

In processes where fast and complex reactions are carried out between unpremixed reactants, process rate and selectivity depend strongly on the way the reacting species are mixed, and therefore on the turbulence conditions in the apparatus.

Several micromixing models based on the turbulence theory have been proposed in the literature. Comparison of experimental results and model predictions have evidenced that relatively simple models assuming a single controlling stage and only one characteristic mixing time can be inadequate if a precise description of the reaction field is required and in addition it may be difficult to assess which characteristic time has to be chosen; on the other side, models that take into account properly the contribution of all the turbulent scales become very complicated and computationally heavy, and in any case the reliability of their predictions is strongly dependent on the accuracy of the hydrodynamic field (Valerio *et al.*, 1994b; Pipino and Fox, 1994; Vanni *et al.*, 1994). Thus, simple models may still be very interesting for engineering applications, but their reliability in complex nonhomogeneous turbulent flows must be assessed.

In this work a tubular reactor with coaxial feeds has been investigated. Experimental hydrodynamic data are not available for this type of device.

Conversion data have been used to calculate the energy dissipation rate by using micromixing models, but the values obtained in this way came out to be significantly higher than those measured on the axis in the case of simple tube flow; this fact lead previous authors to postulate that additional turbulence dissipation takes place (Bolzern *et al.*, 1985; Bourne and Tovstiga, 1988; Bourne and Gablinger, 1989; Bourne and Maire, 1992). It has to be evidenced that in the previous cases a constant value of the dissipation rate  $\varepsilon$  has been assumed, and thus the calculated averaged  $\varepsilon$  value becomes a fitting parameter of the model, in spite of its claimed predictive nature; even when the radial distribution of turbulent energy dissipation is modelled, the centreline value is taken as an adjustable parameter (Bourne and Maire, 1992).

The aim of this work is to evaluate how the average value of the dissipation rate to be employed in monodimensional models is dependent on the operative conditions and on the assumptions of the model and how it is related to the local values evaluated using a CFD code.

## Experimental apparatus and procedures

The set-up is the same described in previous works (Valerio *et al.* 1994a; Pipino and Fox, 1994). The quasi-instantaneous reaction between NaOH and HCl has been adopted as test reaction. The reactants have been fed coaxially at the same average velocity.

The axial concentration profiles have been measured by means of a non-intrusive fiber-optic spectrophotometer. This experimental set-up allows both the evaluation of the local conversion rate

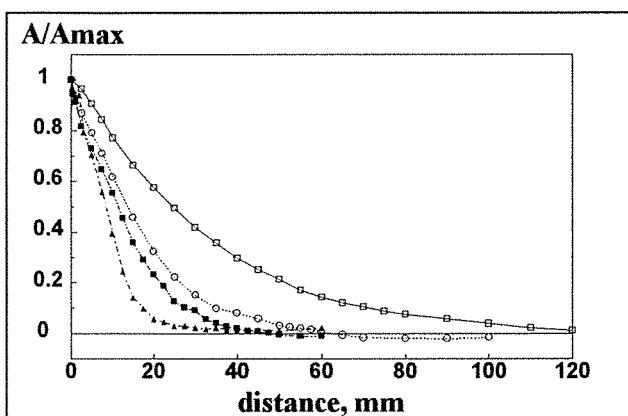


Fig. 1. Axial absorbance profiles for different initial concentration ratios.  $\square$ ,  $c_{A0}/c_{B0}=0.5$ ;  $\circ$ ,  $c_{A0}/c_{B0}=1$ ;  $\bullet$ ,  $c_{A0}/c_{B0}=2$ ;  $\blacktriangle$ ,  $c_{A0}/c_{B0}=20$ . Tube A,  $Re=10,000$ .

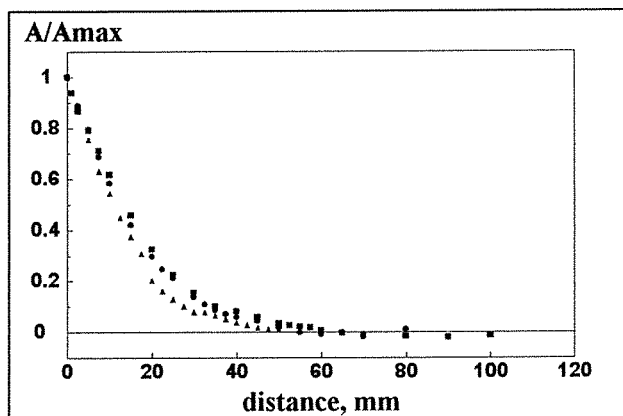


Fig. 2. Axial absorbance profiles for different inner tubes.  $c_{A0}/c_{B0}=1$ ;  $Re=10,000$ .  $\blacksquare$ , tube A;  $\bullet$ , tube B;  $\blacktriangle$ , tube C.

and the reaction time for complete conversion with higher accuracy than the methods based on the visual determination of the length of the reacting plume.

Three different inner tubes have been tested: tube A ( $d_i = 1.17$  mm;  $s = 0.11$  mm), tube B ( $d_i = 1$  mm;  $s = 0.25$  mm) and tube C ( $d_i = 0.72$  mm;  $s = 0.23$  mm). Tubes of different inner diameters,  $d_i$ , and wall thickness,  $s$ , have been chosen, in order to test the effect on the reactor hydrodynamics of both parameters.

Soda initial concentration was kept constant in all the runs (0.01 N) while the acid concentration was varied in order to operate with initial concentration ratios  $c_{A0}/c_{B0}$  in the range 0.5-20 for the three inner tubes. Phenol red has been chosen as indicator, and it has been added to both flows.

Three values of the inlet average velocity were investigated: 1, 1.5 and 2 m/s. In these conditions the outer flow is turbulent while in the inner tube the soda flow is laminar ( $Re = 720-2000$ ); the corresponding Reynolds numbers for the resulting flow, once completely developed, are 10000, 15000 and 20000, respectively.

The absorbance profiles have been made dimensionless dividing them by the maximum absorbance measured at the outlet of the inner tube.

## Results

Axial profiles have been analysed as a function of the three operative parameters ( $c_{A0}/c_{B0}$ ,  $Re$  number and inner tube geometry).

In Figure 1 the dependence of the concentration profiles on  $c_{A0}/c_{B0}$  is shown.

As concerns the effect of the Reynolds number, the dimensionless concentration decreases faster at higher  $Re$  (cf. Valerio *et al.*, 1994a).

In Figure 2 the dependence of the profiles on inner tube geometry is shown. The presence of the inner tube causes an additional turbulent energy dissipation and both the inner diameter and the wall thickness are affecting parameters. First of all, even if the feeds are "isokinetic" in average, each stream has its own velocity profile, and they both have to be modified and to develop up to the velocity profile in the empty tube. This disturbance causes a localised increase in the energy dissipation, that is larger when the internal tube has a larger inner diameter. Moreover, the wall thickness of the inner tube represents a discontinuity zone in the velocity profile: the change in the free section of the tube and the formation of a "wake" on the edge of the inner tube cause additional turbulence.

It has been possible to analyse the two effects more clearly using numerical simulations and the results are given in a further part of the present work.

## Discussion and data interpretation

In the tested reactor, segregation at the scale of the feed tube occurs, so both mesomixing and micromixing must be accounted for.

The predictions of the different models proposed in literature depend dramatically on the value of the turbulent energy dissipation rate. Various authors have observed that the experimental results cannot be interpreted using the Lawn (1971) equation for the evaluation of  $\varepsilon$ ; in fact this equation was experimentally obtained for a simple tubular geometry.

In this work the experimental data were interpreted with two models based on a similar description of diffusion and reaction in a shrinking lamellar system. The first one is the well known EDD model proposed by Baldyga and Bourne (1984a; 1984b) which evaluates the total reaction time  $t_{TOT}$  as:

$$t_{TOT} = 7.5 \left( \frac{d_i}{u} \right) + t_R \quad (1)$$

where the first term on the right hand side of equation (1) represents the life of the initial segregation zone in the form proposed by Bourne and Tovstiga (1988) and  $t_R$  is the reaction time.

The second model is a modified three stage micromixing (TSM) model (Barresi *et al.*, 1992; Pipino *et al.*, 1992). For this model, the total reaction time is obtained as:

$$t_{TOT} = t_{TM} + t_R \quad ; \quad t_{TM} = 0.36 d_i (\nu \varepsilon)^{-1/4} \quad (2)$$

The turbulent mixing time,  $t_{TM}$ , is the time required for the scale of segregation to be reduced up to the point that diffusion and reaction become important. The time of the diffusion and reaction stage,  $t_R$ , is obtained in both cases from the combination of mass-balance equations for each species and the equation of deformation of lamellae. The two models differ substantially in the evaluation of the length of the segregated zone (mesomixing) and in the value assumed for the initial thickness of the laminated structure,  $s_0$ : it is equal to the Kolmogoroff scale,  $\lambda_K$ , in the EDD, and to  $2\pi\lambda_K$  in the TSM model.

Figure 3 shows the dependence of  $\varepsilon$  on  $\bar{u}$  (the mean velocity in the reactor);  $\varepsilon$  has been obtained by fitting the experimental data of the total reaction times.

The interpolation law is nearly the same for the tubes A, B and C.  $\varepsilon$  does not depend on the initial concentration ratio but only on the Re number, as expected; on the contrary, a dependence of the average dissipation rate on the concentration, caused by the variation of the volume of the reaction zone, had been observed by Bourne and Maire (1992).

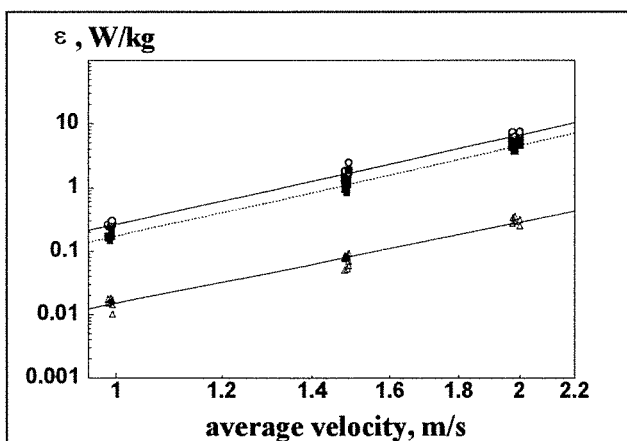


Fig. 3. Average  $\varepsilon$  evaluated from experimental data using the EDD model ( $\Delta$ ) and the TSM model with different initial lamella thickness ( $s_0 = 2\pi\lambda_K$ ,  $\circ$ ;  $s_0 = \pi\lambda_K$ ,  $\blacksquare$ ).

Both models predict that the dependence of  $\varepsilon$  on the turbulence intensity is amplified with respect to what predicted by the Lawn equation as a consequence of the disturbance caused by the presence of the inner tube.

It can be observed that the values of  $\varepsilon$  calculated from the reaction times are very sensitive to the assumptions of the model; this had been already evidenced by Barresi *et al.* (1992). For comparison purposes in Figure 3 the values calculated by the TSM model using different initial thickness of the lamellae are also shown.

The EDD model predicts  $\varepsilon$  values lower than those given by the Three Stage Micromixing model; even larger values of  $\varepsilon$  are obtained if the  $t_{TM}$  and the deformation law are modified in order

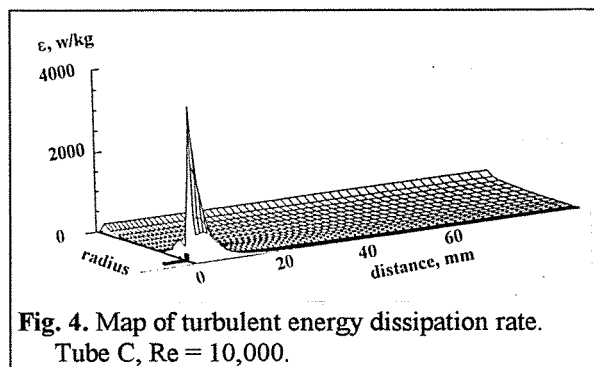


Fig. 4. Map of turbulent energy dissipation rate. Tube C,  $Re = 10,000$ .

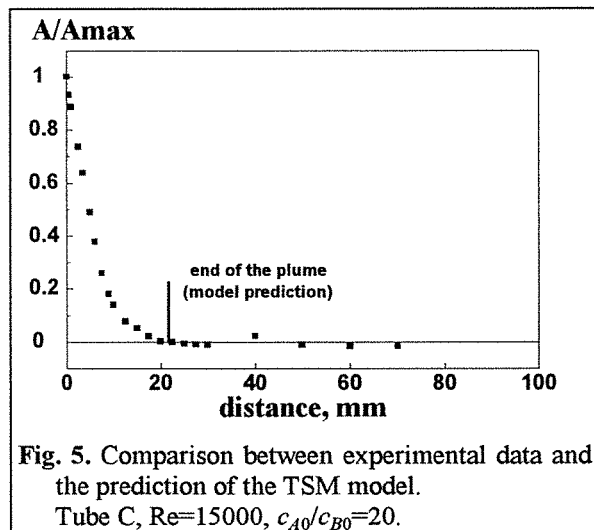


Fig. 5. Comparison between experimental data and the prediction of the TSM model. Tube C,  $Re=15000$ ,  $c_{A0}/c_{B0}=20$ .

to take into account also the inertial effects (Pipino *et al.*, 1993).

This situation is due to the fact that the equation of instantaneous deformation of the lamella used by the models depends strongly on the previous history of the fluid element.

Exponential type deformation laws do not present this disadvantage, but even if, from a theoretical point of view, they appear more correct, do not give satisfactory results when implemented in the previous models.

In this work stronger increases in the values of the dissipation rate have been observed in comparison to that of previous authors (Bolzern *et al.*, 1985; Bourne and Tovstiga, 1988; Bourne and Gablinger, 1989; Bourne and Maire, 1992). This may be due to the difference between the conditions of our experiments and those of the other authors. Reactor scale is very reduced and so disturbance effects, that are very localised, are more important. Moreover, previous works pointed their attention on  $Re$  widely higher than those investigated in our experiments.

To confirm the local influence of the disturbance effects on the turbulent dissipation rate, we simulated the hydrodynamic field of the confined jet reactor with a commercial computational fluid dynamic code

(FLUENT). In this paper, the  $k-\epsilon$  model of Launder and Spalding (1972) has been implemented to evaluate the turbulent quantities. The  $k-\epsilon$  model is an empirical model in which the Reynolds stresses are assumed to be proportional to the mean velocity gradients (Boussinesq hypothesis). An eddy

viscosity that depends on the local turbulent conditions and particularly on the turbulent kinetic energy and turbulent dissipation rate is defined.

Figure 4 shows an example of the turbulent dissipation rate distribution in the reactor. It can be seen that the greatest part of the turbulent dissipation rate occurs close to the edge of the inner tube, due to the recirculation effects, and into the laminar flow, due to the change of the hydrodynamic regime. The relevance of these effects increases at higher Reynolds numbers. They are greatly influenced by the inner tube diameter, while the dependence on the wall thickness is weak. In particular, the dissipation that occurs close to the edge of the inner tube increases for higher tube diameters and decrease for smaller wall-thicknesses.

In Figure 5 a comparison between experimental results and the prediction of the TSM model is shown.

In Figure 6 the comparison between model predictions and experimental data for the three inner tubes at the same  $Re$  is shown. Shorter reaction times are observed in C than in A and B at the same conditions, as a consequence of the

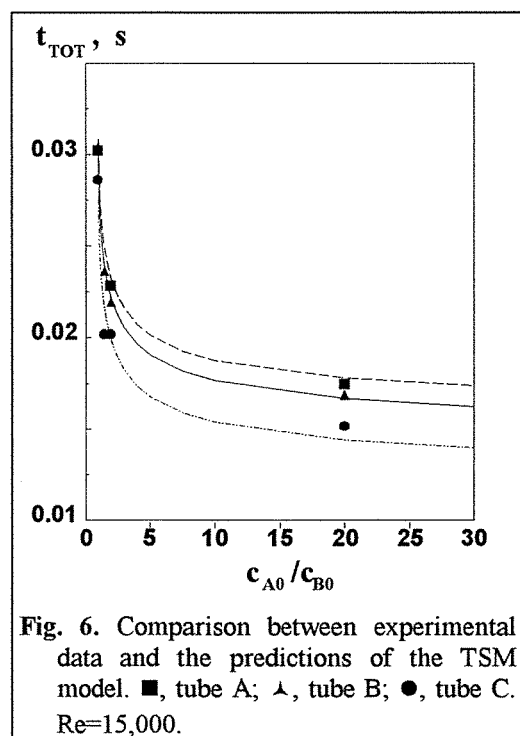


Fig. 6. Comparison between experimental data and the predictions of the TSM model. ■, tube A; ▲, tube B; ●, tube C.  $Re=15,000$ .

geometric characteristics discussed above.

## Conclusions

Experimental conversion data of an almost instantaneous reaction in a tubular reactor with coaxial feeds are presented. The full axial absorbance profile has been measured, to reduce the uncertainty in the evaluation of the length of the reaction plume and to investigate the behaviour in the zone close to the inlet.

The average turbulent energy dissipation rate has been evaluated using both the EDD model and a model previously proposed by the authors, and correlated to the operative conditions. It has been confirmed that the coaxial feed system is a source of additional turbulence dissipation; the simulations have evidenced that the values of  $\varepsilon$  evaluated from the total reaction time are very sensitive to the assumptions of the model: in particular the initial thickness of the lamellar system is a highly speculative parameter. If the value of this quantity is obtained through best fitting of experimental data, obviously different models can give similar performances.

The local distribution of the dissipation rate in the reactor has been simulated using computational fluid dynamics; very strong and localised variations have been evidenced.

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